



IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

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In re the Application of:

Minoru WAKI et al

Group Art Unit: 1755

Serial Number: 09/916,316

Examiner: Callie E. Shosho

Filed: July 30, 2001

For: WATER-BASED INK COMPOSITION FOR INK-JET PRINTING, INK-JET PRINTING METHOD USING THE SAME AND PRINTED MATTER

DECLARATION UNDER 37 CFR 1.132

Commissioner for Patents
Washington, D.C. 20231

Sir:

We, Kazuaki Watanabe and Minoru Waki, residing at c/o SEIKO EPSON CORPORATION, 3-5, Owa 3-chome, Suwa-shi, Nagano-ken, Japan and c/o MIKUNI SHIKISO KABUSHIKI KAISHA, 101, Kokubunji, Mikunino-cho, Himeji-shi, Hyogo-ken, Japan, respectively, duly declare and say:

1. That Kazuaki Watanabe received the degree of Master of Engineering Science from Osaka University, Osaka, Japan, in the year 1987, and has been employed by SEIKO EPSON CORPORATION since 1987;

2. That Minoru Waki graduated from Himeji Institute of Technology, Hyogo, Japan, in the year 1978, and has been employed by MIKUNI SHIKISO KABUSHIKI KAISHA since 1978;

3. That we have read and are familiar with the above-identified patent application and the Office Action thereto mailed July 18,

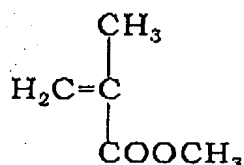
2003;

4. That we made experiments in order to show that an ink composition for ink-jet printing containing as a dispersant a combination of a methyl methacrylate-butyl acrylate-acrylic acid copolymer (I) and a styrene-maleic anhydride copolymer (II) has excellent performances as compared with an ink composition containing a combination of a methyl methacrylate-butyl acrylate-acrylic acid copolymer (I) and a styrene-maleic acid copolymer.

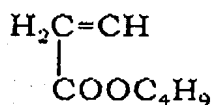
5. The experiments were made as follows:

The following monomers were used in the experiments.

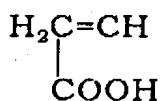
(A) Methyl methacrylate



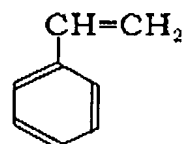
(B) Butyl acrylate



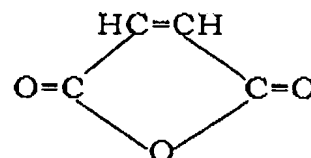
(C) Acrylic acid



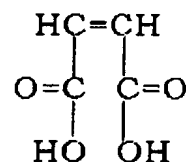
(D) Styrene



(E) Maleic anhydride



(F) Maleic acid



Preparation of copolymers

(a) Methyl methacrylate-butyl acrylate-acrylic acid copolymer (I)

A reactor was charged with a solution of 250 g of methyl methacrylate (A), 82 g of butyl acrylate (B), 30 g of acrylic acid (C) and

0.67 g of azobisisobutyronitrile in 350 g of benzene. After purging the reactor with a nitrogen gas, the solution was subjected to thermal polymerization at 60°C for 7.5 hours with stirring. The reaction product was dissolved in 3 liters of methyl ethyl ketone and precipitated with n-hexane in an amount of 7 times the volume of the resulting methyl ethyl ketone solution to give a copolymer having an acid value based on carboxyl group of 65. The copolymer was dried, pulverized and dissolved in water by neutralization with ammonia water to give a 20 % by weight aqueous solution of copolymer (I) as a dispersant (a).

(b) Styrene-maleic anhydride copolymer (II)

A reactor was charged with a solution of 225 g of styrene (D), 132 g of maleic anhydride (E) and 0.54 g of azobisisobutyronitrile in 300 g of benzene. After purging the reactor with a nitrogen gas, the solution was subjected to thermal polymerization at 70°C for 6.0 hours with stirring and subsequently the resulting copolymer was subjected to half-esterification with n-butanol. The reaction product was dissolved in 3 liters of methyl ethyl ketone and precipitated with n-hexane in an amount of 5 times the volume of the resulting methyl ethyl ketone solution to give a copolymer having an acid value based on carboxyl group of 164. The copolymer was dried, pulverized and dissolved in water by neutralization with monoethanolamine to give a 20 % by weight aqueous solution of half-esterified copolymer (II) as a dispersant (b).

(c) Styrene-maleic acid copolymer (II'-1)

A reactor was charged with a solution of 225 g of styrene (D), 132 g of maleic acid (F) and 0.54 g of azobisisobutyronitrile in 300 g of benzene. After purging the reactor with a nitrogen gas, the solution was subjected to thermal polymerization at 70°C for 6.0 hours with stirring.

The reaction product was dissolved in 3 liters of methyl ethyl ketone and precipitated with n-hexane in an amount of 5 times the volume of the resulting methyl ethyl ketone solution to give a copolymer having an acid value based on carboxyl group of 277. The copolymer was dried, pulverized and dissolved in water by neutralization with monoethanolamine to give a 20 % by weight aqueous solution of copolymer (II'-1) as a dispersant (b1).

(d) Styrene-maleic acid copolymer (II'-2)

A reactor was charged with a solution of 225 g of styrene (D), 79 g of maleic acid (F) and 0.54 g of azobisisobutyronitrile in 300 g of benzene. After purging the reactor with a nitrogen gas, the solution was subjected to thermal polymerization at 70°C for 6.0 hours with stirring. The reaction product was dissolved in 3 liters of methyl ethyl ketone and precipitated with n-hexane in an amount of 5 times the volume of the resulting methyl ethyl ketone solution to give a copolymer having an acid value based on carboxyl group of 166. The copolymer was dried, pulverized and dissolved in water by neutralization with monoethanolamine to give a 20 % by weight aqueous solution of copolymer (II'-2) as a dispersant (b2).

Preparation of ink composition and test thereof

A pigment shown in Table 1 and a dispersant shown in Table 2 were mixed according to the recipe shown in Tables 1 and 2, and the concentration of the dispersed pigment in the resulting mixture was adjusted to 25 % by weight by adding water to the mixture. The mixture was mixed with glass beads (diameter: 1.0 mm, amount: 1.5 times the volume of the mixture) for 2 hours in a sand mill made by Yasukawa Seisakusho Kabushiki Kaisha. After removing the glass beads, to the

mixture were added 20.0 % by weight of glycerol, 7.0 % by weight of maltitol, 2.0 % by weight of 2-pyrrolidone, 1.0 % by weight of an adduct of ethylene oxide to 2,4,7,9-tetramethyl-decyne-4,7-diol (average number of moles of addition:10) and 0.9 % by weight of triethanolamine. The resulting mixture was stirred at ordinary temperature for 20 minutes, and filtered with a membrane filter having a pore size of 10 μ m to give an aqueous ink-jet ink composition.

The volume average particle size of the pigment dispersed in the ink composition was measured to determine the presence of coarse particles having a volume average particle size of 500 nm or more. Also the storage stability, discharge stability and adhesion of the ink composition were measured by the methods described in the specification of the present application.

The results are shown in Table 3.

6. It is observed in Table 3 that ink compositions of Run Nos. 1 to 3 according to the present invention wherein the copolymers (I) and (II) are used together, have excellent storage stability, discharge stability and adhesion. In contrast, comparative ink compositions of Run Nos. 4 to 9 wherein the copolymer (I) is used with a styrene-maleic acid copolymer (II') are much inferior in storage stability and discharge stability as compared with the ink compositions according to the present invention.

Table 1

Run No.	Pigment	
	Kind	Content (% by weight)
1	Pigment Black 7	2.5
2	Pigment Yellow 74	3.5
3	Pigment Red 122	3.0
4	Pigment Black 7	2.5
5	Pigment Yellow 74	3.5
6	Pigment Red 122	3.0
7	Pigment Black 7	2.5
8	Pigment Yellow 74	3.5
9	Pigment Red 122	3.0

Table 2

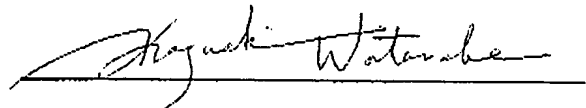
Run No.	Dispersant			Amount based on pigment (% by weight)
	Copolymer (I)	Copolymer (II) or (II')	(I)/(II) or (I)/(II') (weight ratio)	
1	dispersant (a)	dispersant (b)	67/33	25
2	dispersant (a)	dispersant (b)	67/33	25
3	dispersant (a)	dispersant (b)	67/33	25
4	dispersant (a)	dispersant (b1)	67/33	25
5	dispersant (a)	dispersant (b1)	67/33	25
6	dispersant (a)	dispersant (b1)	67/33	25
7	dispersant (a)	dispersant (b2)	67/33	25
8	dispersant (a)	dispersant (b2)	67/33	25
9	dispersant (a)	dispersant (b2)	67/33	25

Table 3

Water-based ink composition for ink-jet printing					
Run No.	Dispersed pigment		Properties		
	Volume average particle size	Presence of coarse particles	Storage stability	Discharge stability	Adhesion
1	122 nm	no	A	A	B
2	145 nm	no	A	A	A
3	139 nm	no	A	A	A
4	167 nm	yes	D	B	A
5	186 nm	yes	D	C	A
6	160 nm	yes	E	B	A
7	131 nm	no	C	B	B
8	153 nm	no	C	C	A
9	147 nm	no	D	C	A

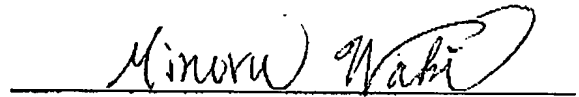
The undersigned declares further that all statements made herein of his own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Date: November 11, 2003



Kazuaki Watanabe

Date: November 10, 2003



Minoru Waki

We, the undersigned witnesses, hereby acknowledge that Kazuaki Watanabe and Minoru Waki are personally known to us and did execute the foregoing Declaration in our presence on:

Date: November 11, 2003 Witness



Date: November 10, 2003 Witness

